

**INVESTIGATION OF PHASE AND EMULSION BEHAVIOR, SURFACTANT
RETENTION, AND CONDENSATE RECOVERY FOR
CONDENSATE/WATER/ETHANOL MIXTURES**

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Investigation of Phase and Emulsion Behavior, Surfactant Retention, and Condensate Recovery
for Condensate/Water/Ethanol Mixtures

ABSTRACT

This semi-annual technical progress report describes work performed at Morehouse College under DOE Grant No. DE-FG26-02NT15447 during the period April 01, 2005 to September 30, 2005 which covers the sixth six months of the project. Presently work is in progress to characterize phase and emulsion behavior for ethylbenzene/water/ethanol system. Ethylbenzene that has the equivalent carbon number is used as the model condensate. In the last reporting period, electrical conductivity measurements for bottom/top, and top/bottom conjugate pair phases of the ethylbenzene/water/ethanol system were performed for various ethanol volume percentage in the mixture: 2,10,20,33,43,50, and 56. During this reporting period, prediction of electrical conductivity data obtained in the past was conducted employing a theoretical model already developed in this project. Results of the comparisons for 2, and 10% ethanol volume in the mixture are presented here. A good agreement was obtained between the predicted emulsion conductivities and the measured values. To date about 99% of the proposed work has been completed. Conductivity prediction for 56 % ethanol volume in the mixture is in progress. Following this prediction, a final report will be developed describing the research activities conducted through the entire project period including results and conclusions.

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INTRODUCTION

The Department of Energy has been sponsoring fundamental and applied research in the natural gas recovery area. Recently, NPTO/Tulsa has been interested in research focusing on innovative methods for technologies towards improved gas and condensate recovery. If an effective method can be found, the potential for incremental recovery is great.

The major factors causing hydrocarbon losses in a reservoir during production of gas and gas condensate fields are related to pressure depletion, retrograde condensation, and water encroachment. The most widespread and simple method of producing gas and gas condensate fields is by depletion, utilizing the natural reservoir pressure only. The major disadvantage of the method is the low condensate recovery. The ultimate condensate recovery from gas condensate fields is 30 - 60 %, depending on the initial content of condensate in gas. In comparison, the final recovery of gas in dry gas fields can be up to 95 %. Despite the low recovery of condensate associated with natural depletion, this method is still widely used in the majority of gas condensate fields in the world. The reasons for this are both technological and economic. Achieving increased production in partially depleted gas and gas condensate fields is essential for many gas producing companies. Incremental production can be very profitable especially in industrially developed countries. Worldwide, the potential for production and improved recovery from gas condensate fields is considerable. In order to improve the recovery from gas condensate fields innovative methods are required.

Depletion of reservoir pressure leads to a reduction in flow capacity and normally causes retrograde condensation. Offsetting the reduced flow capacity by lowering the bottom hole flowing pressure can result in near wellbore retrograde condensation, which leads to decreased effective permeability and additional reduction of flow rates. Lower production rates leads to earlier abandonment resulting in lower ultimate recovery. Significant amounts of condensate can accumulate in the near wellbore zone due to the large pressure drop in this zone. This condensation takes place in addition to the condensation within the reservoir caused by the general pressure depletion. As a result of this condensate accumulation, liquid saturations in the near wellbore zone can considerably exceed the average value over the reservoir as a whole. This causes a large reduction in well productivity. The reduction of well productivity caused by near wellbore accumulation of retrograde condensate has been observed at many fields in the USA, Russia, and Canada. At these fields the well productivity decrease reached 50% in some productive intervals due to condensate accumulation. Further pressure reduction down to the pressure of maximum condensation results in increased volume of condensed liquid. Then the mixture enters a range of direct evaporation and will result in further decrease of pressure. Pressure depletion below the dew-point pressure causes retrograde condensation throughout the entire system that leads to greater than 50% reduction in well productivity.

Water encroachment into a gas or gas condensate reservoir has two opposite effects: 1) maintenance of reservoir pressure, and 2) trapping gas and condensate behind the water level at higher pressure than by depletion and thus reducing the recoverable reserves. Selective water encroachment in a reservoir and breakthrough of water to production wells result in an accumulation

of water in wells and their premature shut-in at higher formation pressures. The impact of each of the above mentioned factors on production and ultimate recovery of condensate and gas varies from reservoir to reservoir, but water encroachment is observed at all gas condensate fields to a certain degree. Application of surfactant flooding technology that is used in the Enhanced Oil Recovery technology may hold great potential for the improved recovery of condensate from the water encroached gas and condensate fields. For this, a research project aimed at understanding the emulsions formed by water, condensate, and a cosurfactant such as ethanol would provide valuable information including how to reduce the interfacial tension between encroaching water and condensate, and under what conditions and how this can be used towards improved recovery. The use of ethanol as the surfactant in this project would also provide valuable information on how to remove the condensate from near the wellbore by reducing the interfacial tension between the gas and condensate and hence improve the gas flow from condensate accumulated wellbores.

Application of surfactants and hence formation of emulsions depend on the desired morphology, for example, a "condensate-in-water" or a "water-in-condensate" emulsion. Formation of the "wrong" morphology or inversion from the desired morphology to the "wrong" morphology causes the product or process to fail. Mixtures of water, condensates, amphiphiles, and salt may separate into three coexisting liquid phases within a well defined temperature interval, the mean temperature of which depends sensitively but systematically on the nature of the condensate and the amphiphile and on the salt concentration. At the mean temperature of this interval one finds--for thermodynamic reasons--a maximum of the mutual solubility between water and condensate and a minimum of the interfacial tension between the aqueous and the condensate-rich phase. Both of these properties are of considerable interest for both research and industrial application.

Many, if not most, of the two-phase emulsions that may commonly exist at various temperatures in systems of surfactant, condensate, and water form in systems that also can form [1] triconjugate condensate-rich top phase (T), water-rich bottom phase (B), and middle-phase microemulsion (M). For such systems, two-phase emulsions may form in any of the various two-phase regions [2-3] that surround a stack of tie triangles in the triangular-prismatic phase diagram. For temperatures between T_{lc} and T_{uc} --the range over which phases T, M, and B coexist--there are three two-phase regions and thus six nonmultiple, two phase emulsion morphologies [4]. For such systems the plotting of phase boundaries and of boundaries between different emulsion morphologies in accordance with the phase rule introduces a rigorous way of showing how the occurrence of each of the six two-phase morphologies depends on temperature and composition. For the limiting tie lines (i.e., the sides of the tie triangle), where the three-phase and two-phase regions meet, the six two-phase morphologies will be formed by the triconjugate phases taken pairwise: T/M, M/T; B/T, T/B; and M/B, B/M [5].

One of the most widely observed phenomena of emulsion inversion is inversion hysteresis [6]. For example, the phase volume fractions at which the inversions A/B to B/A and B/A to A/B occur are somewhat different. If the pair of phase volume fraction points at which the pair of conjugate inversions occur are measured at different temperatures, the resulting measurements form a pair of inversion hysteresis lines in phase volume fraction-temperature space. The fractional phase

volume at which inversion occurs is believed to depend on many different factors, such as the temperature, structure and concentration of the emulsifying amphiphile, salinity, and hydrocarbon composition. The factors that control dispersion morphology and inversion are now well understood [5]. Qualitatively, dispersion inversion has long seemed to be a catastrophic event. The properties of an A/B (A-in-B) dispersion are very different from the properties of a B/A (B-in-A) dispersion, and the inversion from one morphology to the other is often abrupt and irreversible. Inversion is marked by a sudden change in the physical properties (e.g., electrical conductivity) of the dispersion, indicating an abrupt change in the structure of the dispersion [5]. Smith et al. [5,7-10] demonstrated for several oil/water/surfactant systems that elementary catastrophe theory provides a unifying framework to explain these phenomena in systems of variable oil/water ratio and amphiphile concentration at constant temperature and pressure. Smith et al. [1] hypothesized that wherever a phase critical point occurs, there will be a "critical point" for dispersion inversion, also; that is, pairs of dispersion inversion hysteresis lines meet at a point, and this point coincides with the critical point for the two phases of the dispersion.

Sampath et al. [5] determined the morphologies and phase volume fractions at which inversion occurred for (macro)emulsions formed by the middle-phase microemulsion (M) and water-rich bottom phase (B) by means of electrical conductivity measurements for the chemical system $C_6H_{13}(OC_2H_4)_2OH/n$ -tetradecane/aqueous 10mM NaCl at temperatures from 25 °C down to 12 °C, near the lower critical end-point temperature ($T_{lc} = 10.4$ °C). The M/B and B/M morphologies and their inversion hysteresis lines were found to conform to the previously postulated dispersion morphology diagram; that is, within experimental uncertainties, the two emulsion inversion lines in phase volume-temperature space met at a "critical point" that coincided with the lower critical end point for the phases.

The phase behavior, interfacial tension and solution physical parameters only define a single interface in the condensate recovery process, the water-condensate interface. In a condensate and water filled reservoir system, two other interfaces exist, the condensate-rock and water-rock interface. The effect of these two interfaces on the condensate recovery process are just as important, if not more important, than the water-condensate interface [11-14]. Linear and radial corefloods are used to test the effect of the two rock interfaces on the condensate recovery potential of the surfactant systems. Linear corefloods provide adsorption coefficients of chemical onto the rock surface and define fluid rheology as the ability of the solution to flow through porous media. Radial corefloods to estimate condensate recovery potential are preferable to linear corefloods for a number of reasons [11-16]. The pore volume of a radial core is greater than that of a linear core making measurement of the injected and produced fluids volumes more accurate. The flow patterns in a radial core allow expansion of the interface between the injected surfactant and the fluid in the core to occur in a similar manner as in a vertical well in the field. When fractional pore volumes of chemical are injected, chemical banks thin in a manner analogous to a vertical well in the field allowing dispersion, dilution, and other potentially deleterious effects to occur. Recently, Moeti, Sampath, Smith, and Pitts [17-19] investigated the suitability of hybrid (ionic and nonionic) surfactants for enhanced oil recovery and observed inversion hystereses but lower surfactant and hydrocarbon recovery for these surfactants.

To this end, applying our experimental and modeling capabilities in the emulsion research [5,17-19], this project seeks to characterize the phase and morphology behavior of ethylbenzene that has the equivalent carbon number of condensate when mixed with water and a cosurfactant such as ethanol and develop a database for use in improved condensate recovery from water encroached wells. Detailed information on optimal salinity, temperature, emulsion morphologies including inversion hystereses, lower and upper critical points, and recovery efficiency will be obtained for ready use in actual field operations. The information to be obtained will also be useful in evaluating the use of ethanol as the surfactant to remove the condensate from near the wellbore to improve the gas flow in condensate accumulated wellbores.

The specific objectives are:

- 1) Perform salinity and temperature scans to obtain optimal salinity and temperature at which all phases coexist.
- 2) Perform emulsion morphology and coreflooding experiments for the ethylbenzene/water/ethanol system that gives the best results in objective 1.

Experiments and analyses are being carried out to meet the project objectives. To date, salinity and temperature scans of objective 1 were accomplished by Morehouse. Coreflooding experiments were completed by Surtek, our industrial partner in this project. Emulsion morphology experiments were completed by Morehouse in the last reporting period. Prediction of the emulsion data is conducted during this reporting period. The successful accomplishment of the objectives provides a detailed and unique database for use in improved condensate recovery from water encroached wells.

EXECUTIVE SUMMARY

In this semi-annual report, the work performed under DOE Grant No. DE-FG26-02NT15447 during the period April 01, 2005 to September 30, 2005 which covers the sixth six months of the project is described and the major accomplishments are highlighted summarizing the most important research results.

Many surfactants will simultaneously form multiple coexisting liquid phases when mixed with condensate and water within a well defined salinity and temperature interval. At the mean temperature of this interval one finds--for thermodynamic reasons--a maximum of the mutual solubility between water and condensate. In a three phase mixture, the middle phase, which contains the greatest concentration of surfactant, is called a microemulsion. Typically the microemulsion reduces the interfacial tension between hydrocarbon and water by about three or four orders of magnitude. These ultralow tensions allow the hydrocarbon-rich (top) phase to be pushed through porous media by the aqueous (bottom) phase, and the three liquid phases flow as macroemulsions. The microemulsion phase can also form two-phase, or three-phase, macroemulsion structures or morphologies when mixed with either one, or both, of the other two phases, depending on the temperature, relative volumes of the phases, and other factors. Formation of a wrong morphology or inversion from the desired morphology to the wrong morphology is also possible. Investigation of these morphologies, when they occur, and their flow through porous media are of considerable interest for both research and industrial application.

This project seeks to characterize phase and emulsion behavior, and condensate recovery for condensate/water/ethanol systems. The successful accomplishment of the objectives will provide information on the role of salinity, temperature, condensate recovery, and inversion hysteresis of the conjugate emulsion phases for this chemical system. This information will be useful for incremental hydrocarbon recovery during the late period of the production of gas and gas condensate fields.

Presently work is in progress to characterize phase and emulsion behavior for ethylbenzene/water/ethanol system. During this reporting period, prediction of electrical conductivity data obtained in the past was conducted employing a theoretical model already developed in this project. Results of the comparisons for 2, and 10% ethanol volume in the mixture are presented here. A good agreement was obtained between the predicted emulsion conductivities and the measured values. To date about 99% of the proposed work has been completed. Conductivity prediction for 56 % ethanol volume in the mixture is in progress. Following this prediction, a final report will be developed describing the research activities conducted through the entire project period including results and conclusions.

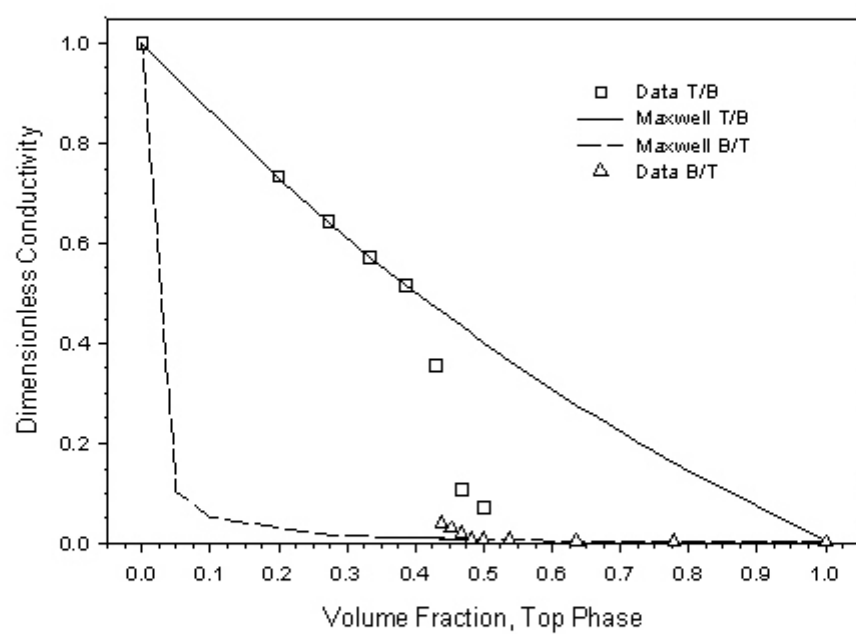


Figure 1. Conductivities for Emulsions of the Bottom and Top Phases formed by 2 vol% Ethanol in Ethylbenzene/Ethanol/aqueous 10 mM NaCl Mixture

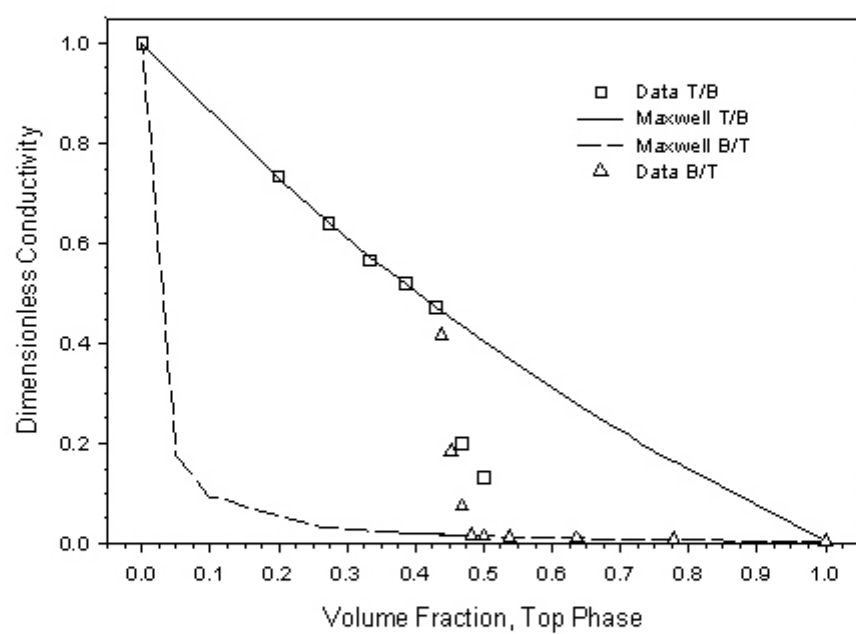


Figure 2. Conductivities for Emulsions of the Bottom and Top Phases formed by 10 vol% Ethanol in Ethylbenzene/Ethanol/aqueous 10 mM NaCl Mixture

EXPERIMENTAL

Presently work is in progress to characterize phase and emulsion behavior for condensate/water/ethanol system.

Phase work including salinity and temperature scans were accomplished by Morehouse in the previous reporting periods. The best ethylbenzene/water/ethanol system defined by the phase work was studied for emulsion morphologies in the last reporting period. Briefly, electrical conductivities were measured for pre-equilibrated top and bottom phases of the mixtures formed with various volume fractions of ethanol (2,10,20,33,43,50,56, and 60%) keeping the volumes of ethylbenzene and water the same in each mixture. Also electrical conductivities were measured for top/bottom, and bottom/top emulsions that form the sides of the tie triangle diagram by mixing measured volumes of the pre-equilibrated phases.

During this reporting period, measured conductivity data were predicted employing a theoretical model (based on Maxwell equations) already developed in this project.

RESULTS AND DISCUSSION

In the last reporting period, using the emulsion measurement system, electrical conductivities were measured for top and bottom phases of the ethylbenzene/water/ethanol system, and for emulsions by mixing measured volumes of the top phase into a fixed volume of the bottom phase and vice versa at room temperature. Electrical conductivity of the emulsion decreased as the fraction of volume of the top phase was increased and vice versa. More experiments in smaller incremental volume of the conjugate phase into the emulsion were performed and the inversion phenomena observed. These experiments were then repeated for top/bottom and bottom/top conjugate pair phases obtained by mixing various ethanol concentrations (2,10,20,33,43,50,56, and 60 vol%) while keeping the volumes of ethylbenzene and water the same in each mixture. Electrical conductivity of the bottom phase decreased as ethanol volume fraction of the mixture increased. Conductivity of the top phase was found small and remained almost the same for variations in ethanol volume fraction of the mixture. Prediction of the emulsion data obtained using the theoretical model already developed in this project was conducted in the current reporting period. Electrical conductivity measurements were then compared with the predictions. Results of the comparisons for 2, and 10% ethanol volume in the mixture are presented in Figures 1 and 2. As can be seen, a good agreement was obtained between the predicted emulsion conductivities and the measured values. To date about 99% of the proposed work has been completed. Conductivity prediction for 56 % ethanol volume in the mixture is in progress. Following this prediction, a final report will be developed describing the research activities conducted through the entire project period including results and conclusions.

Outcome to-date

1. Sampath, R., Dixon, R., Moeti, L., and McCulloch, H., Investigation of Phase and Emulsion Behavior, Surfactant Retention, and Condensate Recovery for Condensate/Water/Ethanol Mixtures, UCR/HBCU/OMI Annual Contractor Review Meeting sponsored by NETL/DOE, June 3-4, 2003, Pittsburgh, PA
2. Sampath, R., Dixon, R., Moeti, L., and McCulloch, H., Investigation of Phase and Emulsion Behavior, Surfactant Retention, and Condensate Recovery for Condensate/Water/Ethanol Mixtures, Spring Seminar Series, Department of Physics and Dual Degree Engineering, Morehouse College, Atlanta, GA, April 2004.
3. Sampath, R., Dixon, R., Moeti, L., and McCulloch, H., Salinity and Temperature Scans for Condensate/Water/Ethanol Mixtures, SPE/DOE Fourteenth Symposium on Improved Oil Recovery, April 20, 2004, Tulsa, OK.
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CONCLUSION

The project is progressing well. To date, phase work including salinity and temperature scans, emulsion morphology measurements, and linear coreflooding experiments were completed. During this reporting period, emulsion morphology data in the form of electrical conductivity responses were predicted using the theoretical model already developed in this project. More data analysis and development of the final report are in progress.

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